Interactions between elastomer fillers, strain, surface properties and friction

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In elastomeric materials, fillers are very important ingredients. There is a wide variety in terms of materials and sizes of the fillers, depending on the application of the specific compound. In a recent study regarding the simulation of the lubricated friction of a rubber material, it became apparent that the fillers can also have a significant influence on the surface properties of a material in combination with mechanical loading. These interactions can be crucial for the correct prediction of the material behavior. It will be shown, how the fillers influence the surface properties of an elastomer, what the most important parameters are and how this can influence the friction properties of the rubber compound.

1 Introduction

Rubber materials for seals are typically designed for a certain range of applications or media and often vary to a very high degree between e.g., O-rings, U-cups or Radial Shaft Seals (RSS). Not only the base polymer (FKM, NBR, EPDM, etc.) but also the fillers change from application to application regarding material (carbon black, minerals, etc.) and size. Furthermore, each rubber material contains a specific set of additives (e.g., anti-ageing, ozone-protection), these are not covered here. In the context of this publication, the term “filler” refers to solid, particle-like filler materials in rubber compounds.

The behavior of the materials needs to be characterized and input in simulations, which are part of the development process of new seals. This also includes mixed friction tribological simulations which are used more frequently recently due to the availability of rubber contact mechanics models (e.g., [1]). In the process of developing such models the validation is very important, i.e., the comparison of the simulation results to other simulation methods and, most importantly, experiments.

However, in this process, one should also question the experimental methods used if an agreement between simulation and experiment cannot be achieved as will be shown in the following chapters.

2 Background

At Freudenberg Technology Innovation (FTI), a mixed lubrication friction model has been developed in the last couple of years. It makes use of an elasto-hydrodynamic (EHL) lubrication model combined with a rubber contact mechanics model [1], [2].

Aside of oil viscosity and material (stress-strain) data, the roughness of the counterparts is a key input parameter for the model to calculate the amount of solid body contact using contact mechanics.

For the validation of the model, Stribeck-curves were measured for RSS-materials sliding against a rotating ground disc (Pin-on-Disc). However, the simulations did not quite match the experiments in the mixed friction regime, while good agreement was achieved in the (elasto-) hydrodynamic regime (see figure 1).

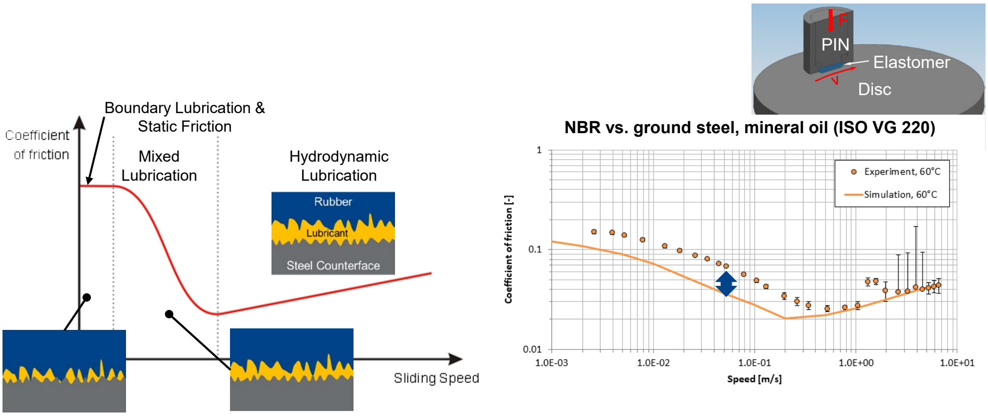

Figure 1: Setup and discrepancy between simulation and experiment

3 Analysis

3.1 White Light Interferometry (WLI)

Taking a closer look at the simulations and the experiments, it was soon discovered that the elastomer samples seemed to be rougher visually after installation in the pin holder for the experiments. This had not been considered before and WLI roughness measurements revealed a surprising amount of increase in roughness: More then 100% in terms of Ra (figure 2).

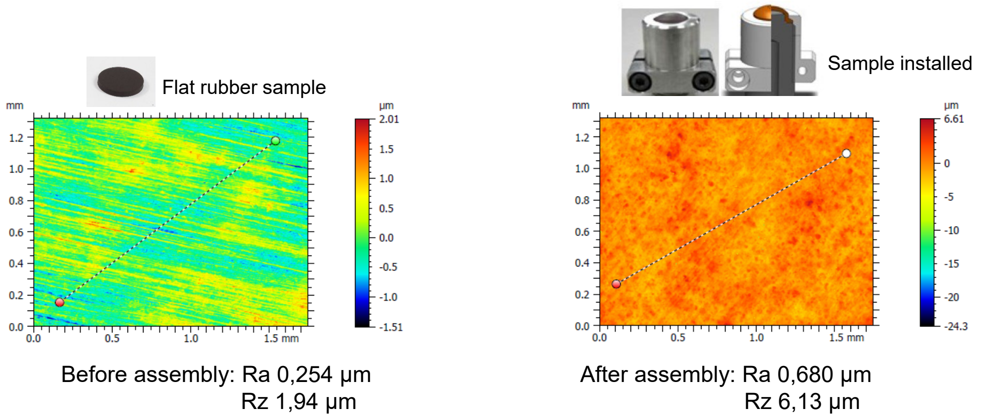

Figure 2: Roughness of NBR sample before and after installation

FEA calculations of the rubber installed in the pin showed that the sample is stretched by 20-30 % at the top surface after installation. To analyse if the phenomenon is purely due to strain, additional materials were selected, and the roughness was measured at different uni-axial strain values. The results indicate a clear dependence on the fillers: The NBR and FKM material have bigger fillers and show a comparably big increase in roughness (about factor 5 at 100% strain for the root mean square roughness Sq), while the EPDM with smaller carbon black particles showed only a small increase and the unfilled EPDM showed no increase (figure 3). The strain of the materials was reduced back to 50% after strain to 100% and the second values at 50% strain indicate that the observed effect is largely reversible when the strain is removed.

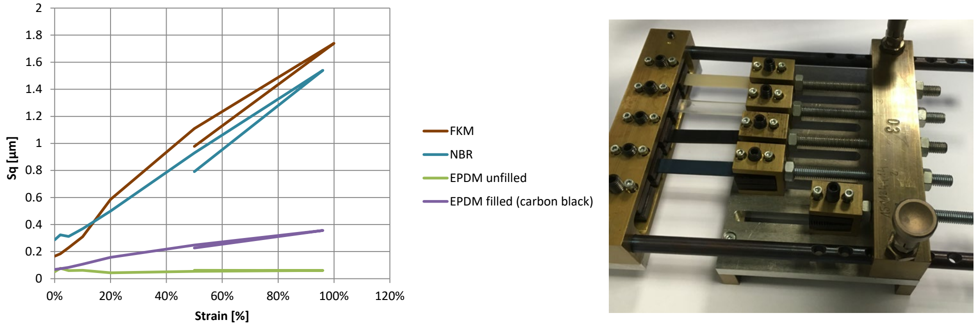

Figure 3: Uniaxial setup and Sq roughness vs. strain

3.2 SEM Microscopy

Using SEM microscopy, the change in surface structure can also be clearly visualized (figure 4). It becomes apparent that under strain, the filler particles dominate the roughness.

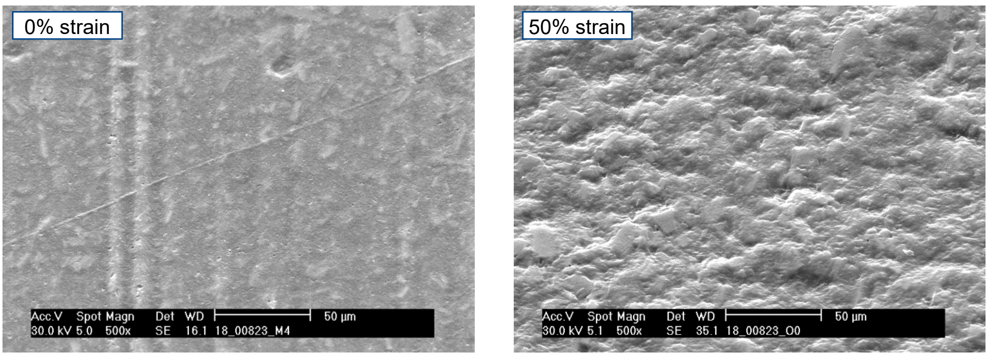

Figure 4: Rubber surface (FKM) with and without strain

Furthermore, in some cases, holes between the filler and the matrix could be observed. This indicates that the rubber is locally detached from the fillers in strained condition (figure 5). It is expected that this is also happening inside the bulk of the rubber.

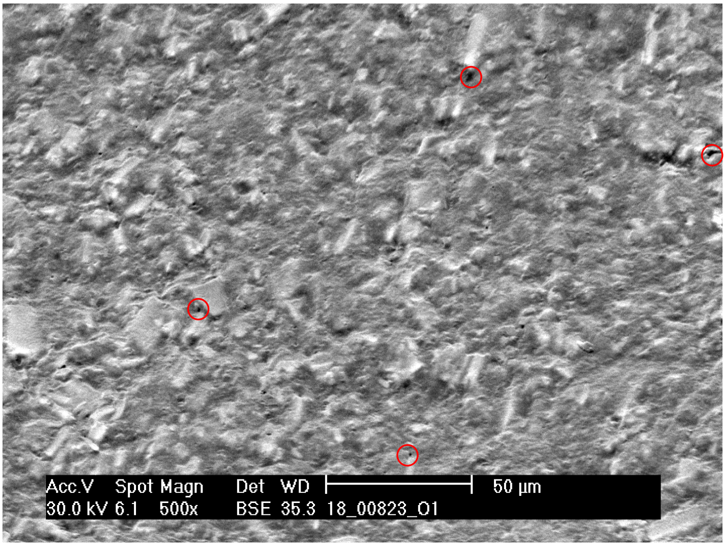

Figure 5: Cracks/holes in the rubber surface at the filler-matrix boundary

3.3 Filler Variation

To understand the effects of the filler only, a special base FKM compound was mixed with different fillers but with the same volume ratio of the fillers. Figure 6 clearly shows that the filler size strongly affects the increase in roughness due to strain but not the initial (as-molded) roughness.

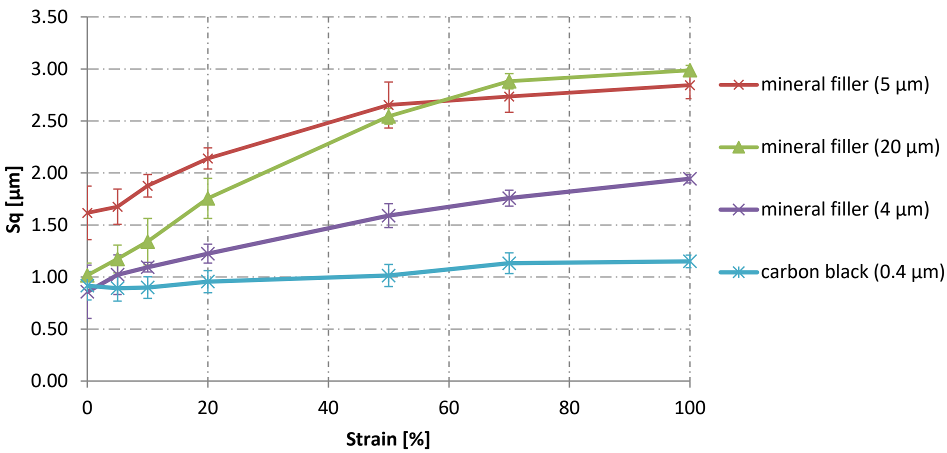

Figure 6: Effect of mean filler particle size on roughness increase

3.4 Simulation

FEA simulations including fillers were also done. In these simulations, the rubber material was filled with spherical particles (10 µm diameter) up to 20 vol. % which were bonded to the matrix and randomly distributed. Other particle shapes and non-bonded fillers proved to be too complex numerically. A representative volume element (RVE) was simulated with a free surface on the top and the bottom of the element (as shown in figure 7) and periodic boundary conditions at the other four surfaces.

The simulations of the RVE show the inhomogeneous stresses between the filler particles and the resulting deformations (figure 7).

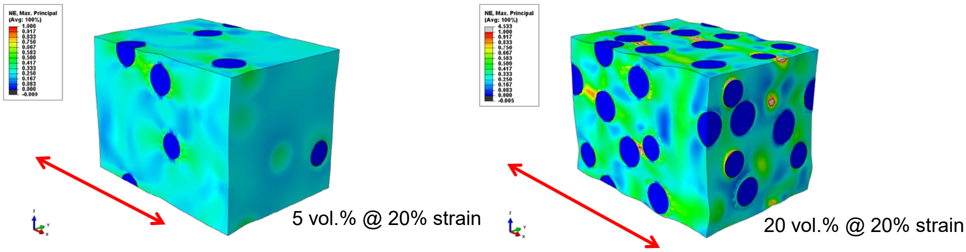

Figure 7: Inhomogeneous strain due to fillers at 20% global strain. The red arrow indicates the direction of strain.

At 20 vol. % filling rate and 20% global strain, locally the strain exceeds 100%, which makes a detachment of the matrix from the fillers very likely in reality. Both, filler amount and strain are not uncommon in sealing applications.

The resulting free surface of the RVE can be periodically continued according to the boundary conditions to create a surface large enough to be facilitate the use of standard surface roughness analysis. This shows an increase in roughness from Sq = 0 µm to Sq = 0.26 µm at 20% strain, in line with the experimental findings above (figure 8).

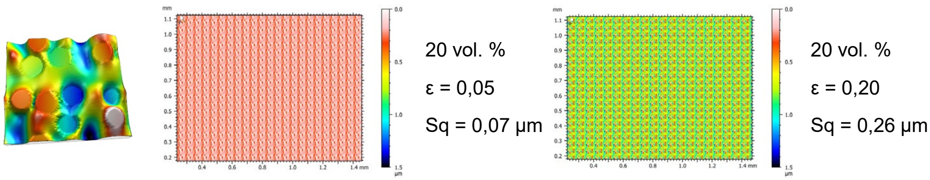

Figure 8: Roughness increase in simulation at 5 and 20% strain (20 vol. % filler ratio)

4 Summary and Conclusion

The effect of roughness increase due to strain of filled elastomers was discovered and analyzed using several techniques. No literature was found on this particular topic, therefore it is assumed to be unknown, at least the extend to which the roughness can increase for commercial rubber compounds (up to 500% at 100% strain!).

It was shown that the increase is directly linked to the filler particles, particularly their size. Hard particles embedded in a soft matrix cause strong stress and strain inhomogeneities which lead to inhomogeneous deformation also at the surface, hence causing the observed changes.

While the general mechanism seems to be quite clear, there are some unanswered questions. A clear relation between filler size and roughness increase was found, but the increase for different volume ratio of fillers (10 and 20 vol. %) showed no clear differences, however the simulations with 5 and 20 vol. % fillers indicated a dependence of the roughness on the amount of fillers. Furthermore, as can be seen from figures 3 and 6, both, linear and decreasing increase of roughness with strain were observed. The reasons for this are not clear, but different relaxation times and material properties could play a role.

In conclusion, this effect might not be relevant for many rubber products but there are certainly applications for which it needs to be considered. In the case of the Pin-on-Disc simulation shown above, unsurprisingly, the agreement with the experiment is very good if the correct roughness data is used for the simulations (figure 9).

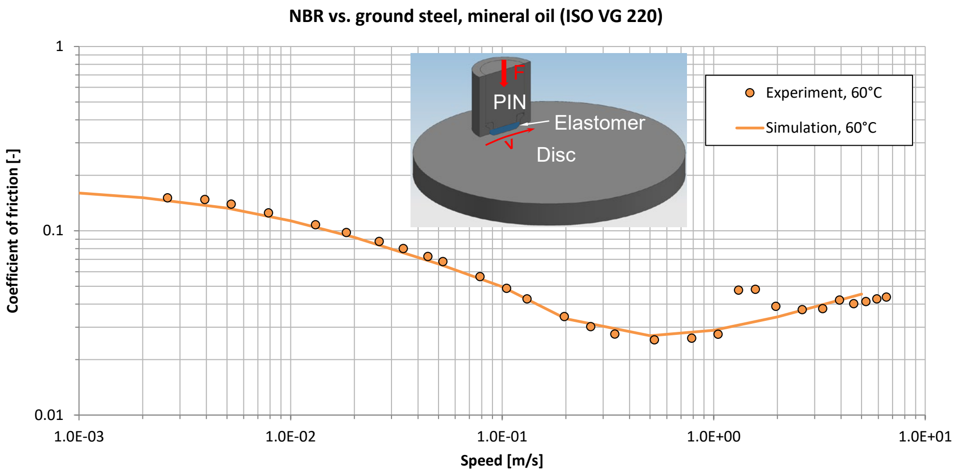

Figure 9: Comparison of experimental results with simulations taking into account the increased roughness of the rubber sample

5 References

[1] Yang, C., Persson, B.N.J.: *Contact mechanics: contact area and interfacial separation from small contact to full contact*, J. Phys.: Condens. Matter, 2008 (20)

[2] Kaiser, F., Gropp, A., Bactavatchalou, R., Rieder, A.: *Simulation of Rubber Friction: Comparison to Pin-On-Disc Measurement Results*, 22nd International Colloquium Tribology, Esslingen, 2020

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